

2003年 3月14日 /時54分

(株) クフレ 知財財産部 葛原

No. 0/09 P. 3

#19  
Ineda  
4/1/03

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION OF :  
TSUYOSHI KOWAKA ET AL. : GROUP ART UNIT : 1713  
SERIAL NO : 09/577,158 :  
FILED: MAY 24, 2000 :EXAMINER: D. R. WILSON  
FOR: POLYVINYL ALCOHOL POLYMER PRODUCTION  
METHOD AND POLYVINYL ALCOHOL POLYMER

Honorable Commissioner of Patent and Trademarks  
Washington, D.C.20231

Sir:

**DECLARATION UNDER 37 CFR 1.132**

I, Kazunori WATANABE, a citizen of Japan, residing at Mizue 170,  
Kurashiki-shi, Okayama-ken, Japan do hereby declare as follows:

I am a co-inventor in the above-identified application.

I graduated from Doctor Course of Faculty of Engineering, Kyushu  
University in March, 1977.

I entered Kuraray Co.,Ltd. in April, 1977, and engaged in the

2003年 3月14日 7時55分

(株) クレ 知的財産部 倉敷

No. 0709 P. 4

development of chemical plants.

I conducted under my supervision and direction the following experiments in order to make clear the present invention.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Further deponent saith not.

March 12, 2003

Date: March 12, 2003

Kazunori Watanabe  
Kazunori WATANABE

### Reference Experiment

Polyvinyl acetate with a limiting viscosity of 1.4 dl/g was dissolved in DMSO and methanol to prepare a paste with a DMSO/polyvinyl acetate ratio of 70/30 (weight ratio) and a methanol mole ratio of 2.55. Sodium methoxide was added at a mole ratio of 0.002 with respect to the vinyl acetate units, and the mixture was loaded into a kneader type mixer (SI Mixer Type SMLJ40, made by Sakura Plant Co., Ltd.) under a normal pressure and a temperature of 100°C to carry out the primary saponification reaction. Upon analyzing the residual acetate groups in the reaction solution by alkalimetry, the degree of saponification was determined to be 93 mole %.

The reaction solution obtained by the primary saponification reaction was then supplied to the tower top of a packed tower filled with Raschig rings and the secondary saponification reaction was carried out under a pressure of 550 Torr and a reaction temperature of 100°C while distilling off the methyl acetate that was produced and the unreacted methanol. The final degree of saponification attained was 97 mole % and the average degree of polymerization of the PVA that was obtained was 1720. The reaction solution obtained by the saponification reaction was a solution with a PVA concentration of 19.5 wt%; and this solution could be used as it is as a spinning stock solution in a spinning process to produce fiber. The block character value of the PVA was

0.99.

### Experiment 1

Deeming the saponification reaction solution obtained in Reference Experiment to be the first stage saponification reaction solution, this reaction solution was used as the second stage saponification stock solution to carry out the saponification reaction in two stages. For the primary saponification reaction of the second stage, the same kneader type mixer as that used for the first stage of the Reference Experiment was used and the saponification reaction was carried out under a pressure of 4.0kg/cm<sup>2</sup>G and a temperature of 95°C. The degree of saponification attained was 99.2 mole %. This primary saponification reaction solution was supplied in an up-flowing manner to the lower part of a shell and tube evaporator and the secondary saponification reaction of the second stage was carried out under the conditions of a pressure of 50 Torr and a temperature of 90°C. The final degree of saponification attained was 99.9 mole % and the average degree of polymerization of the PVA was 1720. The reaction solution obtained by the saponification reaction was 1 weight % content of methanol and a PVA concentration of 20 weight %, and this solution could be used as it is as a spinning stock solution in a spinning process to produce fiber. The block character value of the PVA was 0.99.

## Experiment 2

The saponification was carried out in the same manner as the Experiment 1, except that the same packed tower as the secondary saponification reaction of the first stage of the Reference Experiment was used for the secondary saponification reaction of the second stage. The final degree of saponification attained was 99.9 mole % and the average degree of polymerization of the PVA that was obtained was 1720. The reaction solution obtained by the saponification reaction was a solution with a PVA concentration of 20.0 wt%, and this solution could be used as it is as a spinning stock solution in a spinning process to produce fiber. The block character value of the PVA was 0.99. But, the reaction solution of the secondary saponification reaction of the second stage was high viscous. Therefore, a special reboiler was necessary for carrying out the secondary saponification reaction of the second stage by the packed tower.

## Experiment 3

The saponification was carried out in the same manner as the Experiment 2, except that the same shell and tube evaporator as the secondary saponification reaction of the second stage of the Experiment 1 was used for the secondary saponification reaction of the first stage. The final degree of saponification attained was 99.2 mole % and the

2003年 3月14日 7時56分

(株) クラレ 知的財産部 倉敷

No. 0709 P. 8

average degree of polymerization of the PVA that was obtained was 1720. The reaction solution obtained by the saponification reaction was a solution with a PVA concentration of 20.0 wt%, and this solution could be used as it is as a spinning stock solution in a spinning process to produce fiber. The block character value of the PVA was 0.99.

#### Experiment 4

The saponification was carried out in the same manner as the Experiment 3, except that the same shell and tube evaporator as the secondary saponification reaction of the first stage was used for the secondary saponification reaction of the second stage. The final degree of saponification attained was 99.2 mole % and the average degree of polymerization of the PVA that was obtained was 1720. The reaction solution obtained by the saponification reaction was a solution with a PVA concentration of 20.0 wt%, and this solution could be used as it is as a spinning stock solution in a spinning process to produce fiber. The block character value of the PVA was 0.99.



Saponification reaction					Results
The first stage		The second stage			
The primary reactor	The secondary reactor	The primary reactor	The secondary reactor		
Exp.1	A kneader mixer	A packed tower	A kneader mixer	A shell and tube evaporator	Good
Exp.2	A kneader mixer	A packed tower	A kneader mixer	A packed tower	A complicated reboiler is needed
Exp.3	A kneader mixer	A shell and tube evaporator	A kneader mixer	A packed tower	A degree of saponification of 99.6 mole % or more can not be achieved
Exp.4	A kneader mixer	A shell and tube evaporator	A kneader mixer	A shell and tube evaporator	A degree of saponification of 99.6 mole % or more can not be achieved

TC 1700 MAIL ROOM

MAR 31 2003

RECEIVED